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# On-board and Off-board performance of hydrogen storage options for light-duty vehicles

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## ARTICLE INFO

### Article history:

Received 19 November 2010

Received in revised form

4 April 2011

Accepted 6 May 2011

Available online 15 June 2011

### Keywords:

Hydrogen storage

Compressed hydrogen

Liquid hydrogen

Metal hydride storage

Sorption

Hydrogen carriers

## ABSTRACT

Leading physical and materials-based hydrogen storage options are evaluated for their potential to meet the vehicular targets for gravimetric and volumetric capacity, cost, efficiency, durability and operability, fuel purity, and environmental health and safety. Our analyses show that hydrogen stored as a compressed gas at 350–700 bar in Type III or Type IV tanks cannot meet the near-term volumetric target of 28 g/L. The problems of dormancy and hydrogen loss with conventional liquid H<sub>2</sub> storage can be mitigated by deploying pressure-bearing insulated tanks. Alane (AlH<sub>3</sub>) is an attractive hydrogen carrier if it can be prepared and used as a slurry with >50% solids loading and an appropriate volume-exchange tank is developed. Regenerating AlH<sub>3</sub> is a major problem, however, since it is metastable and it cannot be directly formed by reacting the spent Al with H<sub>2</sub>. We have evaluated two sorption-based hydrogen storage systems, one using AX-21, a high surface-area superactivated carbon, and the other using MOF-177, a metal-organic framework material. Releasing hydrogen by hydrolysis of sodium borohydride presents difficult chemical, thermal and water management issues, and regenerating NaBH<sub>4</sub> by converting B–O bonds is energy intensive. We have evaluated the option of using organic liquid carriers, such as n-ethylcarbazole, which can be dehydrogenated thermolytically on-board a vehicle and rehydrogenated efficiently in a central plant by established methods and processes. While ammonia borane has a high hydrogen content, a solvent that keeps it in a liquid state needs to be found, and developing an AB regeneration scheme that is practical, economical and efficient remains a major challenge.

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## 1. Introduction

A number of physical and materials-based hydrogen storage options are being developed to meet simultaneously the vehicular targets for gravimetric and volumetric capacity, cost, efficiency, durability and operability, fuel purity, and environmental health and safety. In this paper, we review the performance characteristics of the currently available physical, metal

hydride (MH), sorption, and chemical hydrogen storage methods and assess them for their potential to meet these targets.

Table 1 lists the various storage options examined in this work. The list does not necessarily include all the candidate options, only the ones that we have evaluated to-date. Compressed hydrogen storage at 350–700 bar in Type III (metal-lined) and Type IV (polymer lined) vessels has been demonstrated in a number of prototype fuel cell vehicles.

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**Table 1 – Important attributes of physical and material-based storage systems analyzed in the present study.**

Storage Options	On-Board Storage			Production, Delivery, and Regeneration Infrastructure	Refueling Station
	Peak P	Peak T	$\Delta H$		
	bar	K	kJ/mol-H <sub>2</sub>		
<b>Physical Storage</b>					
Compressed Hydrogen	350–700	300	NM	SMR, Compressors, Pipeline Delivery	Tube Banks, Compressors
Liquid Hydrogen	6–8	20	NM	SMR, Liquefiers, Trucks	LH <sub>2</sub> Storage
Cryo-compressed Hydrogen	50–300	20–50	NM	SMR, Liquefiers, Trucks	LH <sub>2</sub> Storage, Liquid H <sub>2</sub> Pumps
<b>Metal Hydride Storage</b>					
Sodium Alanate	100	423–453	37–47	SMR, Compressors, Pipeline Delivery	Tube Banks, Compressors, Chillers
Alane	10–50	423–523	7	SMR, Two-way delivery, Central Regeneration	Fresh and Spent Fuel Storage
<b>Sorption Storage</b>					
AX-21	50–300	77–120	3–6	SMR, Liquefiers, Trucks	LH <sub>2</sub> Storage, Liquid H <sub>2</sub> Pumps
MOF-177	50–250	77–120	2–5	SMR, Liquefiers, Trucks	LH <sub>2</sub> Storage, Liquid H <sub>2</sub> Pumps
<b>Chemical Hydrogen Storage</b>					
Sodium Borohydride	10–50	423–523	53–63	SMR, Two-way Delivery, Central Regeneration	Fresh and Spent Fuel Storage
N-ethylcarbazole	10–50	423–523	51	SMR, Two-way Delivery, Central Regeneration	Fresh and Spent Fuel Storage
Ammonia Borane	10–50	423–523	20–33	SMR, Two-way Delivery, Central Regeneration	Fresh and Spent Fuel Storage

Compressed hydrogen storage in carbon fiber (CF) wound pressure vessels is well understood, as is the inherent short-coming that since the density of hydrogen is 39 g/L at 700 bar, this option cannot possibly meet the intermediate (2015) system-level target of 40 g/L, after including the volume of the hardware in the system. Although demonstration vehicles have also been built with liquid hydrogen (LH<sub>2</sub>) tanks, the interest in LH<sub>2</sub> storage has subsided because of the energy inefficiency incurred in H<sub>2</sub> liquefaction and an unavoidable loss of H<sub>2</sub> during extended parking, even from tanks with cryogenic multi-layer vacuum insulation (MLVI).

The problem of dormancy and H<sub>2</sub> loss from LH<sub>2</sub> storage tanks can be overcome by using insulated pressure vessels, i.e., cryo-compressed hydrogen storage (CCH<sub>2</sub>). Aceves et al. [1,2] have built three generations of CCH<sub>2</sub> tanks to demonstrate the advantages of CCH<sub>2</sub> systems over compressed and liquid hydrogen systems.

For hydrogen storage in metal hydride (MH) systems, much attention has been focused on sodium alanates as hydrogen storage materials starting with the discovery that they can be made reversible by doping with Ti catalysts [3]. Researchers continue to use NaAlH<sub>4</sub> as a surrogate material in their studies on engineering of MH systems, while recognizing that this material may not be a long-term solution for automotive storage systems [4,5]. Alane (aluminum hydride, AlH<sub>3</sub>) is another metal hydride that has many interesting properties as a potential hydrogen storage material [6,7]. It is a covalently bonded, binary hydride that can exist in several phases, all of which are thermodynamically unstable, but kinetically metastable at room temperature because of their slow decomposition rates. It contains 10.1-wt% hydrogen and has the desired attribute of low heat of formation ( $\Delta H$ ). However, the Gibbs free energy of formation of the stable phase ( $\alpha$ -AlH<sub>3</sub>) at 298 K is 48.5 kJ/mol, making it impractical to regenerate AlH<sub>3</sub> from spent Al with gaseous H<sub>2</sub> on-board a vehicle.

Lightweight, porous, high surface-area sorbents, to which hydrogen bonds only weakly, are being investigated as potential storage materials. In general, the storage capacities of such materials, including the recently developed metal-organic frameworks [8,9], are less than 2 wt% at room temperatures. It is well known that physisorption can be significantly enhanced by operating at cryogenic temperatures [10,11]. Superactivated carbon AX-21 has shown excess adsorption of 5.4-wt% H<sub>2</sub> at liquid nitrogen temperature (77 K) and moderate pressure (<90 bar). MOF-177, Zn<sub>4</sub>O(1,3,5-benzenetribenzoate)<sub>2</sub>, has shown an even higher uptake at 77 K: 7.5-wt% of excess H<sub>2</sub> and 11.2-wt% of absolute H<sub>2</sub> at 70 bar [8]. In this overview, we assess the potential of meeting the gravimetric and volumetric capacity targets for on-board H<sub>2</sub> storage systems with cryo-adsorption on AX-21 and MOF-177 at moderate pressures (up to 350 bar but preferably <100 bar).

On-board storage capacity may be significantly enhanced with the use of hydrogen carriers that would be regenerated off-board (and may suffer an energy penalty in the process). Our overview includes three hydrogen carriers that are representative of the main classes of organic and inorganic chemical hydrides being investigated by the storage community. Sodium borohydride appears attractive as it can release 4 mol of hydrogen per mole of NaBH<sub>4</sub> through the hydrolysis reaction [12]. Some of the disadvantages are that NaBH<sub>4</sub> needs to be carried as a solution with an alkaline additive for stabilization; the hydrolysis reaction is highly exothermic and requires difficult thermal and water management; and converting the product B–O bonds to B–H is energy intensive. N-ethylcarbazole represents a class of liquid organic carriers that can release hydrogen by thermolytic decomposition over a catalyst, and be rehydrogenated off-board by established industrial methods [13]. The challenge has been to find suitable organic carriers that have sufficient hydrogen capacity, optimal  $\Delta H$ , rapid decomposition kinetics and that remain liquid over the working temperature range. Finally, ammonia

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